

Amendments to the Claims

This listing of claims will replace all prior versions, and listings, of claims in the application:

Listing of Claims:

Claims 1-2 (Canceled)

Claim 3 (Currently Amended): The process according to claim 22~~23~~, wherein said non-hydrolyzable ~~substituent group~~ is H, an alkyl, aryl or fluoroalkyl group or an aminoalkyl group.

Claim 4 (Currently Amended): The process according to claim 22, wherein said step e) ~~(f)~~ of drying the gel is a liophilisation carried out at a pressure lower than 70 mm Hg, to obtain a mesoporous aerogel powder.

Claim 5 (Currently Amended): The process according to claim 22, wherein said step e) ~~(f)~~ of drying the gel is a mild heat treatment carried out at an atmospheric pressure and a temperature no greater than 100°C.

Claim 6 (Previously presented): The process according to claim 22, wherein said nitroxyl radical is added

to said solution along with said monomer precursor in a one-step procedure.

Claim 7 (Currently Amended): The process according to claim 22, wherein in a two-step procedure, first said ~~monomer precursor compound~~ that forms 2, 2, 6, 6 - tetramethyl-1-piperidinyloxy under the instant reaction conditions is hydrolyzed in part with water in the presence of an acid and then said nitroxyl radical is added to this solution, to obtain a porous sol-gel polymeric oxide with a fractal macromolecular structure.

Claim 8 (Canceled)

Claim 9 (Previously presented): The process according to claim 22, wherein said reductive amination is carried out by stirring a solution of said 4-oxo-TEMPO in methanol with said 3-aminopropyl-trimethoxysilane, and reducing the thus formed imine with NaBH_3CN .

Claim 10 (Currently Amended): The process according to claim 6, wherein said nitroxyl radical is TEMPO ~~or a precursor thereof~~ or a compound that forms 2, 2, 6, 6 - tetramethyl-1-piperidinyloxy under the instant reaction

conditions and said radical is physically entrapped within a sol-gel matrix adding a solution thereof in methanol to said ~~precursor~~ compound following said one-step procedure.

Claim 11(Previously presented): The process according to claim 22, wherein said catalytic porous materials are in the shape of powders, films, monoliths, or fibers.

Claims 12-13(Canceled)

Claim 14(Previously presented): A process according to claim 23, wherein said liquid phase is an organic solvent, a biphasic organic solvent-water system, or water and said primary oxidant is NaOCl, NaOBr, HNO₃, CuCl/O₂, K₃Fe(CN)₆, or NO₂.

Claim 15(Previously presented): A process according to claim 23, wherein said alcohol substrate is an alkyl alcohol, an aryl alcohol, a steroid alcohol, an allylic alcohol, a terpenoid alcohol or retinol and it is oxidized in a bi-phasic reaction system CH₂Cl₂-H₂O, said primary oxidant is aqueous alkaline NaOCl and wherein said nitroxyl radical is 4-oxy-TEMPO and said monomer precursor is 3-aminopropyl-

trimethoxysilane to obtain a catalytic material containing chemically linked radicals.

Claim 16(Currently Amended): A process according to claim 23, wherein said alcohol substrate is a monomer or an oligomeric carbohydrate protected at the anomeric center, said solvent is water, said oxidant is alkaline NaOCl or NaOCl in the presence of a catalytic amount of NaBr, and wherein said nitroxyl radical is 4-oxy-2, 2, 6, 6 - tetramethyl-1-piperidinyloxy~~TEMPO~~ and said monomer precursor is 3-amino-propyl-trimethoxysilane to obtain a catalytic material containing chemically linked radicals.

Claim 17(Previously presented): A process according to claim 16, wherein said catalytic material is in the form of pumice stones coated with said sol-gel film doped with said nitroxyl radicals, and said carbohydrate is a water soluble polymer.

Claim 18(Canceled)

Claim 19(Currently Amended): The process according to claim ~~22~~23, wherein P is a non-hydrolyzable ~~substituent~~group.

Claim 20 (Canceled)

Claim 21 (Currently Amended): A process according to claim 23, wherein said alcohol substrate is an alkyl alcohol, an aryl alcohol, a steroid alcohol, an allylic alcohol, a terpenoid alcohol or retinol and it is oxidated in a bi-phasic reaction system $\text{CH}_2\text{Cl}_2\text{-H}_2\text{O}$, wherein said primary oxidant is aqueous alkaline NaOCl , and wherein said nitroxyl radical is 4-oxy-2,2,6,6,-tetramethyl-1-piperidinyloxy and said ~~monomer precursor~~ compound that forms 2, 2, 6, 6 - tetramethyl-1-piperidinyloxy is 3-aminopropyl-trimethoxysilane to obtain a catalytic material containing chemically linked radicals, wherein said radical is tethered to said ~~monomer precursor~~ compound that forms 2, 2, 6, 6 - tetramethyl-1-piperidinyloxy through reductive amination by stirring for three hours a solution of 4-oxy-2,2,6,6,-tetramethyl-1-piperidinyloxy in methanol with a slight excess of 3-aminopropyl-trimethoxysilane, and reducing the thus formed imine with NaBH_3CN .

Claim 22 (Currently Amended): A process for the preparation of a reactive sol-gel catalytic porous material comprising chemically doping said material with stable organic nitroxyl radicals, by carrying out the steps of:

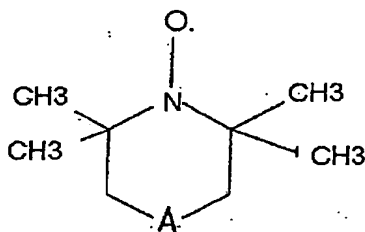
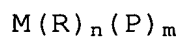
copolymerizing a solution including:

- a) 3-amino-propyl-trimethoxysilane as a monomer precursor;
- b) a dopant consisting of 4-oxy-TEMPO as a stable nitroxyl radical or a ~~precursor thereof~~ compound that forms 2, 2, 6, 6 - tetramethyl-1-piperidinyloxy under the instant reaction conditions;
- c) a solvent including H₂O and a co-solvent selected from the aliphatic alcohols; an acid or base to catalyze the processes of sol-gel hydrolysis and copolymerization; and one or more additives selected from those known to be useful in the preparation of porous materials to form a gel containing said dopant trapped therein;
- d) tethering said radical to said monomer precursor through reductive amination, said solution including H₂O as a solvent and a co-solvent from the aliphatic alcohols; an acid or base to catalyze the processes of sol-gel hydrolysis and copolymerization; and one or more additives selected from those known to be useful in the preparation of porous materials to form a gel containing said dopant chemically trapped therein;
- e) evaporating said solvent;
- f) drying said gel; and

g) coating said gel on a mesoporous inorganic support.

Claim 23(Currently Presented): A process for a liquid-phase oxidative conversion of a substrate of a primary or secondary alcohol into a carbonyl or carboxyl derivative thereof, comprising said conversion being carried out with said substrate in catalytic presence of a reactive sol-gel catalytic porous material either chemically or physically doped with stable organic nitroxyl radicals, said material being coated on a mesoporous inorganic support and containing a copolymer of

a) at least one monomer precursor selected from the group consisting of metal and semi-metal alkoxides, metal esters and semi-metal esters, of the general formula



wherein M is a metal or a semimetal, R is an hydrolyzable substituent, P is a non-hydrolyzable group, n is an integer of 1 to 6, and m is an integer of 0 to 6, and

b) a dopant consisting of a stable di-tertiary-alkyl nitroxyl radical or a precursor thereof of the formula, wherein A represents a chain of two or three carbon atoms, one or two of said carbon atoms being eventually substituted by one oxygen or nitrogen atom, and one or more additives selected from those known to be useful in the preparation of porous materials to form a gel containing said dopant trapped therein.

Claim 24(Previously presented): A process for liquid-phase oxidative conversion of a substrate of a primary or secondary alcohol into a carbonyl or carboxyl derivative thereof, comprising conducting said oxidative conversion in the presence of a doped catalytic material according to claim 22, and in the presence of a primary oxidant effected in selective alcohol oxidations mediated by nitroxyl radicals.

Claim 25(New): A reactive sol-gel catalytic porous material comprising a mesoporous inorganic support and a sol-gel catalytic porous material coated on said mesoporous inorganic support, said sol-gel porous material comprising a copolymer of

a) 3-amino-propyl-trimethyloxysilane as a monomer precursor and

b) a dopant consisting of 4-oxy-2,2,6,6-tetramethyl-1-piperidinyloxy as a stable nitroxyl radical or as a precursor thereof tethered to said monomer precursor through reductive amination, and

c) at least one additive selected from those known to be useful in the preparation of porous materials to form a gel containing said dopant trapped therein.